Nutrient recovery from wastewater treatment by ultrafiltration membrane for water reuse in view of a circular economy perspective

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Abstract

The study aims to recover nitrogen from wastewater by employing ultrafiltration membrane in water reuse for agriculture purpose. To such aim, a new reclaimed water quality index (RWQI) is proposed and applied including an innovative protocol for its assessment. Specifically, the influence of filtration and backwashing times for an ultrafiltration system aimed to nutrient recovery has been analyzed. The final goal was to pin down the trade-off between operation costs and effluent quality. Results show that backwashing time play a crucial role in reducing the operation costs; indeed, low values (i.e., 0.5 min) lead to an increase in the number of required chemical cleanings and consequently operation costs (namely, up to 0.042 €/m^3). The compromise among effluent quality and operation costs has been obtained for 7 min and 1 min, filtration and backwashing, respectively.

Keywords: wastewater treatment, ultrafiltration membrane, fouling, reclaimed water quality index, nutrients recovery

1 Introduction

The forecasted water demand by 2030 is 64% more than the current amount of water available worldwide (Maryam and Büyükgüngor, 2019; Ahmad et al., 2022). Agricultural sector is responsible for 70% of water consumption in the world (FAO, 2021). With the increasing population, agricultural and industrial water footprints increase as well (Wang and Ge, 2020). Furthermore, as a result of climate change, rainfall in dry areas are expected to decrease, consequently alternative water sources are needed (Chen et al., 2021). One way to reduce the water footprint of agricultural sector is to decrease the consumption by applying efficient irrigation strategies such as drip irrigation (Pereire et al., 2009). Another way to reduce the water footprint is to irrigate the fields using treated wastewater (Chen et al., 2021). Treated wastewater is also a reliable water source for areas not traditionally arid but that are facing water crisis due to climate change (Lazarova et al., 2013).

Wastewater is traditionally seen as a waste that requires to be treated before disposal in view of protecting receiving water bodies; however, with the increasing number of areas that face with water crisis, this approach towards wastewater has changed over the years (Yadav et al., 2021). Indeed, wastewater treatment plants (WWTPs) are now being transformed into resource recovery facilities from which recover water, energy, nutrients (phosphorus and nitrogen) and biosolids (Rao et al., 2017).

Depending on the water reuse purpose, reclaimed water quality and thus required treatment level changes (Yang et al., 2020). In order to spread the use of reclaimed water in agriculture, European Parliament published minimum water quality criteria (Regulation 2020/741) for reclaimed water and divided reclaimed water into four quality classes. In these four classes, secondary treatment and disinfection are compulsory. However, treatment degree decreases from the highest reclaimed water class (class A), which has no restrictions of using reclaimed water in irrigation, to the worst (class D), which is restricted to be applied only on commercially processes crops that has no contact with humans or livestock. Suspended solids, organic matter, and pathogens are the main concerns for irrigation water (Fito and van Hulle, 2021). Reclaimed water should have five-day biochemical oxygen demand (BOD_5) and total suspended solids (TSS) concentrations below 10 mg/L, turbidity below 5 NTU, and *Escherichia coli* (*E. coli*) count below 10 cfu/100 mL to be classified as class A. To achieve this, tertiary treatment after secondary treatment of wastewater is needed.

Membrane filtration, especially ultrafiltration (UF), is one of the most used technologies to produce irrigation water from secondary effluent of WWTPs, because it has high TSS, organic matter, and pathogen removal capacity (Oron et al., 2008). However, membrane fouling decreases permeability, thus increasing operation costs due to energy consumption and chemical cleaning frequency (Perez et al., 2022). Chemical cleaning can remove irreversible fouling on the membrane surface that physical cleaning cannot remove (Lin et a., 2010). In order to decrease chemical cleaning frequency, fouling mitigation control strategies such as aeration, employing backwash into filtration cycle, and coagulant addition can be used (Cornelissen et al., 2007; Mannina and Cosenza, 2013). However, these control strategies increase operating costs. A trade-off between operation costs and permeate quality is needed also in view of carbon footprint reduction. Supplying air at a low flow to a membrane reactor decreased operation costs by 20% (Mannina and Cosenza, 2013). On the other hand, high airflow caused reduction in particle size and decreased permeate quality in addition to increased energy cost.

Cornelissen et al. (2007) showed that employing air/water backwashing in filtration cycle is effective to mitigate membrane fouling. Yang et al. (2021) studied optimization of filtration cycle in a real wastewater treating semi-industrial pilot plant to provide minimum reclaimed water quality criteria. They showed that with decreasing backwash frequency increased the irreversible fouling formation and stated the optimum filtration to backwash ratio as 3 and 4 (total filtration cycle was 60 min).

Effluent quality indexes (EQIs) are used in benchmarking studies to determine the effect of different control strategies on the treatment performance of WWTPs (Jeppson et al., 2007). EQIs simplify the comparison of control strategies by providing a single output for each strategy. To calculate the EQI, weight factors are assigned to each effluent quality parameter and then weighted factors are summed up. When different control strategies are analyzed, considering only water quality parameters may lead to misevaluation in water reuse; water quality parameters are focused in treatment performance only. Indeed, in view of a resource recovery perspective nutrients have to be considered as an adding value (i.e., fertilizers) especially when the treated water is reused in agriculture purposes. A new EQI able to take into account the above elements is needed. Such EQI needs to indicates both the undesired and desired parameters' effect on reclaimed water quality according to reuse purpose.

In this study, the effluent of a pilot-scale activated sludge process was treated with UF membranes to produce irrigation water within Wider-Uptake project (Mannina et al., 2021). UF membrane system was operated with a periodic filtration and air/water backwash cycle. To determine the best operation conditions having minimum operation cost, different cycle times and configurations were studied. Four experimental stages were compared in terms of effluent quality and treatment cost. A novel reclaimed water quality index (RWQI) suitable for agricultural reuse was proposed and applied.

2 Materials and Methods

2.1 Pilot plant description

A pilot plant using UF as tertiary treatment for water recycling and reuse (Figure 1) was built-up at the Water Resource Recovery Laboratory of Palermo University (Mannina et al., 2021). The pilot plant was designed for an influent flow rate of 20 L/h and was fed with the effluent of a pre-denitrification activated sludge plant treating the real wastewater collected within the University of Palermo Campus (Mannina et al., 2021). The pilot plant consists of a membrane tank, blower (LA-45C, Nitto Kohki, Germany), filtration and backwash pumps (Qdos30 metering pumps, Watson Marlow, UK), and UF membrane module (Figure 1). Hollow-fiber UF membrane (Koch separation solutions, Wilmington - USA) having 0.03 µm pore size and 1.4 m^2 total surface area was used. The filtration and backwash pumps have 30 L/min capacity. Sodium hypochlorite (NaOCl) was used for membrane chemical cleaning according to the cleaning protocol provided by the manufacturer. Disinfection system consists of a mixing tank (250 mL), contact tank (10 L), and dosing pump. For permeate disinfection NaOCl was dosed at 5 mg Cl_2/L concentration into the mixing tank, then the mixture of permeate and disinfectant flowed through a plug-flow contact tank (contact time was 30 min). Disinfected permeate was stored in the storage tanks having 20 m^3 volume before being used. The average concentrations of TSS, BOD_5, NH_4-N, and PO_4-P in the influent of UF were 34±22 mg/L, 33±13 mg/L, 16±8.9 mg/L, and 5.7±4.3 mg/L, respectively. Turbidity was 1.6±1 NTU and *E. coli* was log 4.4 cfu/100 mL in the UF influent.

2.2 Experimental campaign

Four experimental stages with different ratios of filtration (F) to backwashing (B) times were studied (Table 1). During each stage, the membrane fouling was investigated by continuously monitoring the transmembrane pressure (TMP) and the permeate flow rate. During the experimental campaign, physical (manually) and chemical membrane cleanings were performed to reduce the transmembrane pressure (TMP) according to Chang et al. (2001). In particular, physical (manually) cleanings were performed to reduce the TMP value when increased (as absolute value) till to 0.6 bar; according to the manufacturer suggestions, if the TMP reached the absolute value of 0.8 bar after a physical cleaning a chemical cleaning was performed by using NaOCl.

The frequency of physical and chemical cleaning was recorded to investigate membrane fouling behavior under different ratios of filtration to backwashing time.

2.3 Analytical Methods

TSS, BOD_5, ammonia (NH_4-N) and orthophosphate (PO_4-P) in the feed, permeate and disinfected water were measured two times per week according to Standard Methods (APHA, 2012). For the same samples, *E. coli* were also measured by using method F as proposed by IRSA - CNR (2003). Turbidity was measured by using a portable Hanna (USA) HI93703 turbidimeter.

2.4 Evaluation Criteria

2.4.1 Reclaimed water quality index

To evaluate the reclaimed water quality, two indexes have been here proposed. The first index is the Reclaimed Water Quality Index no.1 ($RWQI_1$) and the second index is the nutrient benefit/drawback RWQI (named $RWQI_2$). $RWQI_1$ (Equation 1) represents the normalized (with respect to the EU reclaimed water quality criteria) weighted sum of the pollutant concentrations contained in the water to be reused.

$$RWQI_1 = \frac{1}{\sum_{i=1}^{n} w_i \, C_{i,c}} \left(\sum_{i=1}^{n} w_i \, C_{i,e} \right) \tag{1}$$

Where, w_i is the weight factor of ith pollutant, $C_{i,c}$ is the concentration of the pollutants stated in the EU criteria (namely, BOD_5 and TSS [kg/m^3], turbidity [NTU], and *E. coli*- [log cfu/100 mL]), and $C_{i,e}$ is the concentration of the pollutants measured in the effluent, and n is the number of pollutants taken into account.

$RWQI_1$ varies between 0 and 1. $RWQI_1$ for the EU criteria is equal to 1. If $RWQI_1$ of the effluent is lower than 1, the water is appropriate for irrigation because the sum of weighted concentrations of pollutants in the effluent is lower than the EU minimum reclaimed water reuse criteria. Therefore, $RWQI_1$ allows to compare the reclaimed water quality with the minimum reclaimed water quality criteria set by EU.

$RWQI_2$ contains the weight factors of $RWQI_1$ for the same pollutants, however it also includes nitrogen and phosphorus as compounds to be considered. Therefore, $RWQI_2$ takes into account the possibilities to use nutrients contained in the effluent as fertilizers in case of water reuse for agriculture purpose. For this reason, it is assumed that the desired compounds for irrigation water are nitrogen and phosphorus.

2.4.2 Calculation protocol for RWQI assessment

In Figure 2, the stepwise (five steps) proposed protocol is reported. The final goal of the protocol is to assess the net $RWQI_2$ which represents the normalized weighted sum of the compounds that are limited by the EU minimum reclaimed water quality criteria together with the compounds that are desired for irrigation water (such as nutrients). The net $RWQI_2$ of each stage is between -1 and 1 (-1 is the most undesired and 1 is the most desired option). As first step, desired and undesired compounds are selected. Such compounds are defined according to water reuse purpose. Specifically, for water reuse in agriculture, desired compounds are selected as nitrogen and phosphorus, while undesired compounds are selected as the compounds (*E. coli*, BOD_5, TSS, NTU) limited in the EU criteria. In second step, the weight factors are assigned. The weight factors for desired compounds are positive and sum to 1. On the other hand, the weight factors for undesired compounds are negative and sum to -1. In third step, the weighted sum of desired ($RWQI_{2,\ desired}$) and undesired ($RWQI_{2,\ undesired}$) compounds are then calculated according to Equation 2. In fourth step, both $RWQI_{2,\ desired}$ and $RWQI_{2,\ undesired}$ are normalized with respect to the maximum sum of the desired and undesired values obtained (see Equation 3). By this way, normalized $RWQI_2$ of all stages are in the range of 0 to 1 for desired compounds and -1 to 0 for undesired compounds. In fifth step, the net $RWQI_2$ can be calculated as the sum between normalized $RWQI_{2,\ desired}$ and normalized $RWQI_{2,\ undesired.}$

$$RWQI_2 = \sum_{i=1}^{n} w_i\, C_i \tag{2}$$

$$Normalized\ RWQI_2 = \frac{1}{(\sum_{i=1}^{n} w_i\, C_i)_{max}} \left(\sum_{i=1}^{n} w_i\, C_i \right) \tag{3}$$

Weight factors for undesired compounds were selected as -0.4 for *E. coli* and -0.2 for BOD_5, TSS and turbidity. For desired compounds of TN and TP the weight factors were 0.5.

2.4.3 The Operation Cost

The operation cost (OC) of the pilot plant was calculated as the sum of the energy cost (C_{energy}) due to the energy consumption of the filtration and backwashing pumps (E_{pump}), the energy required for aeration (E_{aer}), and the chemical costs used in membrane chemical cleaning (C_{chem}). E_{pump} [kW] was calculated according to Equation 4 where TMP is the transmembrane pressure [kPa], Q_{eff} is the effluent flow rate [m^3/h] of the pump, η is the pump efficiency. Equation 5 was used to calculate E_{aer} [kW] where w is the mass flow of air [kg/s], R is the gas constant for air [8.314 kJ/kmol/K], T is the air temperature [K] and e is the blower efficiency (taken as 0.8). The inlet and outlet pressures [atm] are p_1 and p_2, respectively. 29.7 and 0.283 are the constants according to the international unit system and for air, respectively. The unit energy cost (γ_e) used in Equation 6 is taken as 0.28 €/kWh according to the Italian fare.

$$E_{pump} = \frac{1}{t_1 - t_0} \sum_{t=0}^{t=n} \frac{TMP\ Q_{eff}(t)}{100\ \eta} \tag{4}$$

$$E_{aer} = \frac{w\ R\ T}{29.7\ (0.283)\ e} \left(\left(\frac{p_2}{p_1} \right)^{0.283} - 1 \right) \tag{5}$$

$$C_{energy} = \left(E_{pump} + E_{aer} \right) \times \gamma_e \tag{6}$$

$$C_{chem} = N \times \gamma_c \tag{7}$$

3 Results and Discussion

3.1 Average compounds measured

In all the tested ratios of filtration and backwashing times, the UF membrane successfully captured all the parameters limited in the minimum reclaimed water quality criteria. The *E. coli* measured in the permeate was 0 in all stages as expected. The BOD_5 concentrations in the permeate was 4.1 ±0.05 mg/L, 8.9±0.13 mg/L, 5.4±0.54 mg/L, and 6.5±0.09 mg/L, S1-S4, respectively. The TSS concentrations were always below 2 mg/L and turbidity values were below 1 NTU for the permeate of all stages. Similar results were achieved by Gomez et al. (2007) who measured 0 cfu/100 mL of *E. coli* and 1-7 mg/L of TSS in the UF permeate. Moreover, Falsanisi et al. (2010) observed 0 cfu/100 mL of *E. coli*, <0.2 mg/L of TSS, and <0.2 NTU of turbidity in the effluent of secondary treatment filtration by UF membranes. Perez et al. (2022) reported 17 cfu/100 mL of *E. coli* and 1.2 mg/L of TSS in the UF permeate. Mainardis et al. (2022) stated that with UF membrane filtration, bacteria removal up to log 5-6 can be achieved.

3.2 $RWQI_{1\text{-}2}$ assessment

To calculate $RWQI_1$, weight factors were assigned to compounds defined in the criteria (*E. coli*, BOD_5, TSS, and turbidity). The distribution of weight factors was arranged according to the importance of the parameters and the sum of weight factors was equal to 1. *E. coli* was selected as the most important parameter and has the highest weight factor (0.4), because it is the direct indicator of fecal pollution. The weight factors of BOD_5, TSS, and turbidity were selected as equal (0.2).

Figure 3 shows the average $RWQI_1$ for stages S1-S4 compared with the value obtained for the water classified as “class A” according to EU minimum reclaimed water quality criteria (Regulation 2020/741). From Figure 3 one can observe that for all stages, the average $RWQI_1$ was lower than that obtained for class A water. Therefore, by analyzing the results only in terms of $RWQI_1$ one should say that under all the tested stages the quality of the obtained water is excellent according to EU regulation (Regulation 2020/741). In particular, under stage S1 with 2.5 min of filtration and 0.5 min of backwashing time (F/B ratio was 5) the lowest $RWQI_1$ value was obtained (0.26) (Figure 3). Conversely, under stage S2 with filtration time of 6 min and backwashing time of 1 min (F/B ratio was 6) the highest $RWQI_1$ value was obtained (0.44) (Figure 3). However, $RWQI_1$ overlooks the nutrients in the reclaimed water. Robles et al. (2020) stated that using nutrient-rich permeate of membrane treatment in agriculture can be considered as a technology for nutrient recovery. Moreover, Mainardis et al. (2022) showed that the use of nutrient-rich treated wastewater in agriculture can save between 24 and 161 €/ha per year in fertilizer use, depending on crop type. So, inclusion of nutrients in the reclaimed water is significant in scenario evaluation. Therefore, the net $RWQI_2$ of each scenario should be analyzed to select the absolute best one.

Indeed, $RWQI_2$ includes nitrogen and phosphorus in the effluent so the benefit obtained from reclaimed water reuse in agriculture can be compared to determine the best operation of the UF membrane system. The permeate TN concentration for S1, S2, S3, and S4 was 6.6±1.0 mg/L, 13.6±7.6 mg/L, 26.8±2.3 mg/L, and 16.3±2.6 mg/L, respectively. The permeate PO_4-P concentration was 2.3±0.5 mg/L, 2.3±0.2 mg/L, 2.0±0.0 mg/L and 4.1±0.0 mg/L, for S1-S4, respectively. Such a significant difference in soluble pollutants, especially in terms of nitrogen was most likely due to the variations in upstream treatment performance. Nevertheless, the effect of upstream treatment performance was neglected in the present study to prove that considering only undesirable compounds in treatment performance evaluations can lead to error.

Figure 4 shows the results of the average $RWQI_2$ obtained for each stage (S1-S4). The best scenario in terms of net $RWQI_2$ was S3 (total cycle time was 8 min and F/M was 7) since S3 has the highest positive net $RWQI_2$ value indicating that desired compounds are higher than undesired. Indeed, From Figure 4 one can observe that although S1 had the lowest $RWQI_2$ for undesired compounds, it also had the lowest $RWQI_2$ for desired compounds. Since S3 had the highest $RWQI_2$ for desired compounds, the net $RWQI_2$ of S3 was the highest among all scenarios. Therefore, despite in terms of $RWQI_1$ the best result could be S1, when the role of nutrients is included (calculating net $RWQI_2$) S3 become the best result. Consequently, in a situation where all operational scenarios achieve limits, analyzing control strategies by considering only undesirable parameters may result in ignoring the benefits that would have been obtained from desired parameters. Thanks to the net $RWQI_2$ defined in this study, the desired and undesired compounds can be selected in accordance with the water reuse purpose in scenario analysis studies, and the evaluation can be made without overlooking the benefits of water reuse.

3.3 Operation costs

Figure 5 shows the energy consumption of aeration and pumps for each stage. The specific aeration energy consumption of the UF pilot plant was in the range of 0.247 and 0.260 kWh/m^3. The blower capacity was kept identical in all stages. However, ambient temperature changes slightly variated aeration energy consumption. The average ambient temperature during S1 and S2 operation was 14.3℃, while it was 18.5℃ for S3 and S4.

The highest specific pumping energy consumption was observed in S2 as 0.696 kWh/m^3 and the lowest was observed in S4 as 0.429 kWh/m^3. Perez et al. (2022) estimated UF pump specific energy consumption as 0.18 kWh/m^3 for filtration of 104 L/h secondary treatment effluent. Kehrein et el. (2021) reported the specific operation cost (that includes energy, chemical, maintenance, membrane replacement, and labor costs) of UF membranes (0.1 L/h treatment capacity) as 1 kWh/m^3. This difference with respect to the literature is mainly due to the capacity of the treatment plant. Indeed, with the increase of the treatment capacity specific cost of treatment plants decreases (Judd and Judd, 2006).

Specific operation cost of each scenario is shown in Figure 6. Total specific energy cost varied between 0.203-0.274 €/m^3, while total specific chemical cost was in the range of 0.029-0.060 €/m^3. The specific chemical cost increase was aligned with the increase of the filtration time in S2, S3, and S4. In terms of chemical consumption, S2 was the most favorable operation scenario with the lowest specific chemical cost (chemical cleaning applied 1 time in 10 days of operation) (0.029 €/m^3). The lowest specific operation cost was estimated for S3 as 0.236 €/m^3 because it had the lowest specific energy cost (0.203 €/m^3). On the other hand, the highest specific operation cost was belonged to S4 (0.334 €/m^3). Perez et al. (2022) assumed unit energy cost as 0.11 €/kWh and estimated UF filtration's cost of energy and chemical consumption as 0.131 €/m^3. In the present study, specific energy cost estimation was higher than Perez et al. (2022) study because unit energy cost was assumed as 0.28 €/kWh according to Italian energy tariff (EC, 2020).

Comparison of operating scenarios showed that S3 with 7-minute filtration time and 1-minute backwash time had the lowest operating cost and highest $RWQI_2$. If nitrogen and phosphorus were not taken into account (as in $RWQI_1$), the operation scenario with the lowest operating cost would still have to be chosen as S3 optimum, as all operation scenarios meet minimum recovered water quality criteria. However, the use of $RWQI_2$ has brought S3 forward in terms of irrigation water quality in addition to cost.

4 Conclusions

To determine the best operation condition that provides optimal water quality for irrigation and had minimum treatment cost, different filtration and backwashing times were experimentally studied. The minimum reclaimed water quality criteria were achieved in all scenarios. A novel water quality index, $RWQI_2$, was introduced as a tool to compare different treatment scenarios for water reuse purposes. Scenario evaluation showed that S3 with 7-minute filtration and 1-minute backwash time had the lowest operating cost and highest $RWQI_2$. Increasing filtration time increased chemical consumption. The highest energy cost was observed in the scenario with the longest filtration time suggested by manufacturer.

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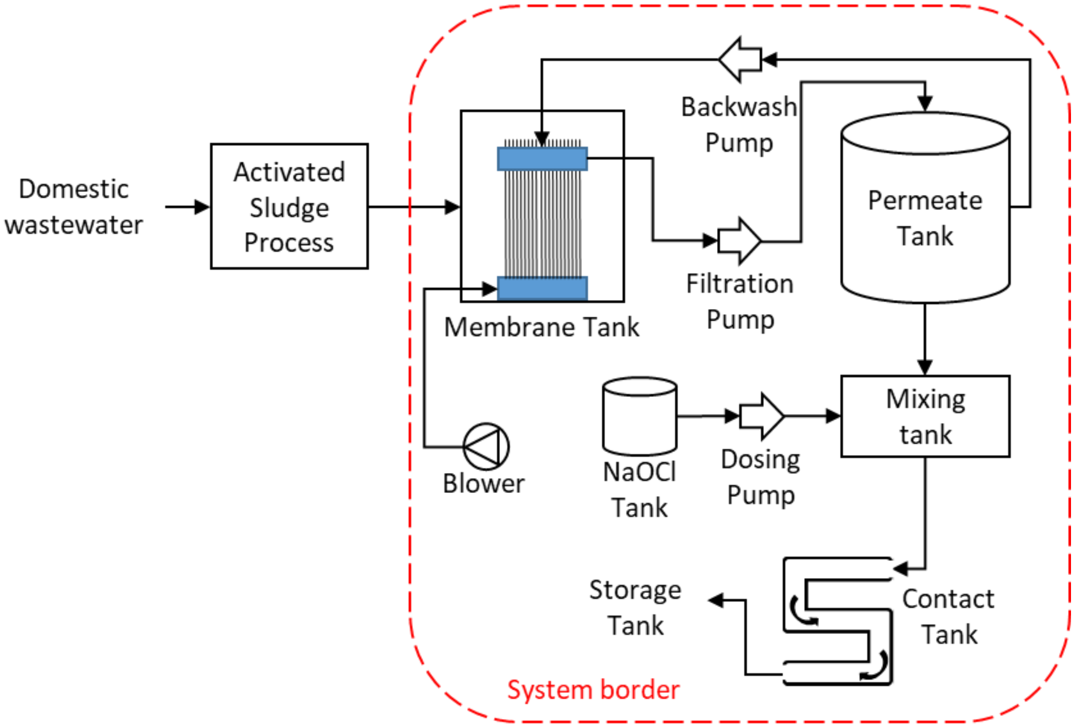

Figure 1. Experimental set-up

Table 1. Experimental stages of the study

	Unit	**Stage 1 (S1)**	**Stage 2 (S2)**	**Stage 3 (S3)**	**Stage 4 (S4)**
Filtration time (F)	min	2.5	6	7	9
Backwashing time (B)	min	0.5	1	1	1
Total cycle time	min	3	7	8	10
F/B ratio	-	5	6	7	9

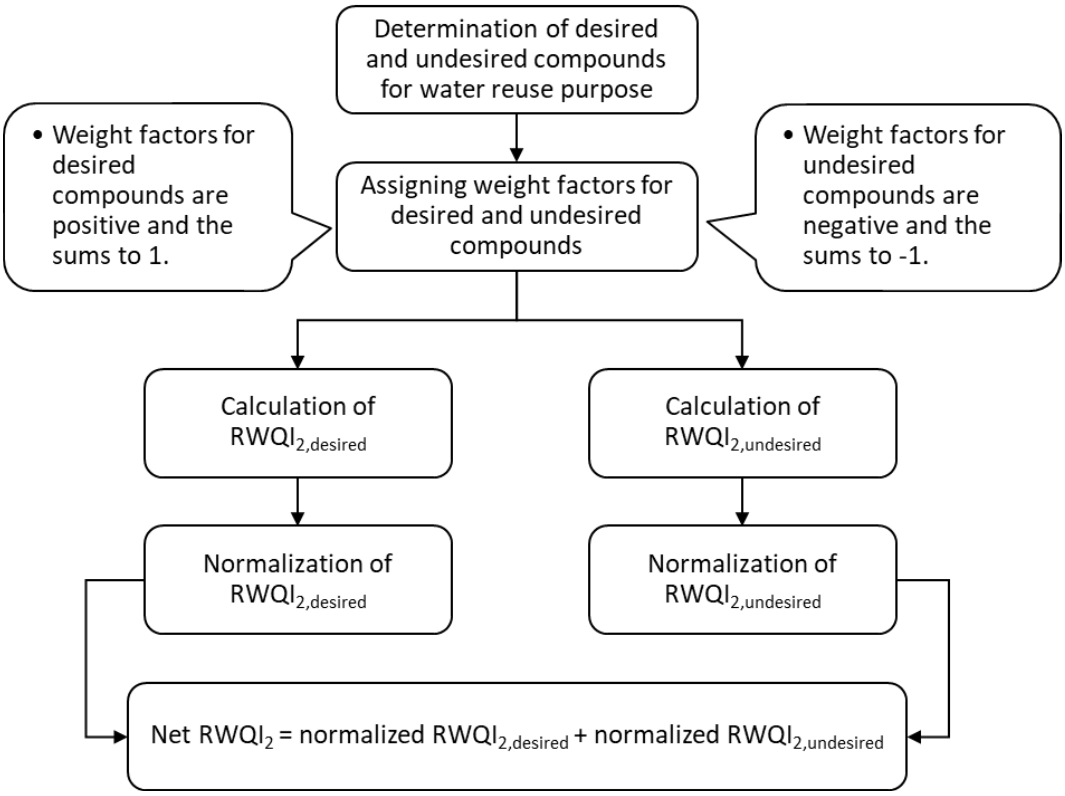

Figure 2. $RWQI_2$ calculation steps

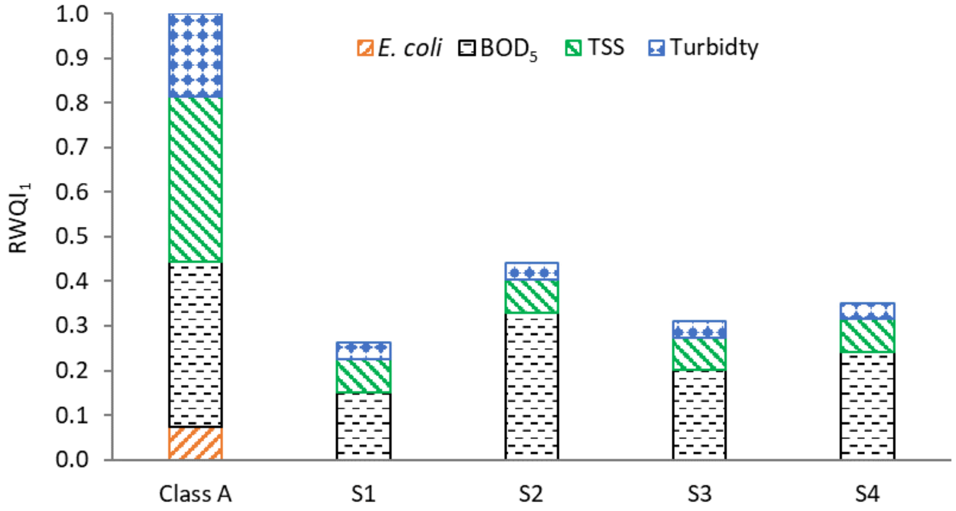

Figure 3. Comparison of reclaimed water quality index of each scenario and minimum reclaimed water quality criteria Class A

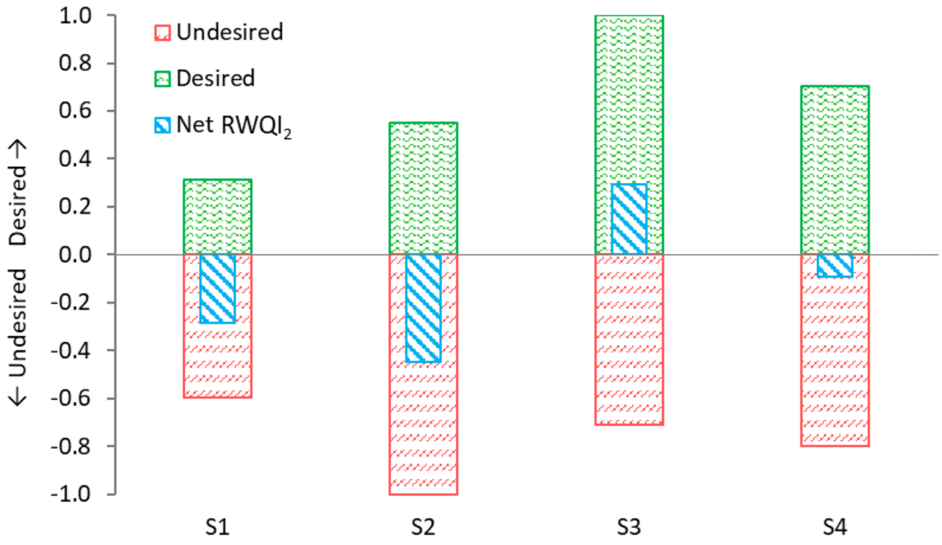

Figure 4. Comparison of RWQI$_2$ of operation stages/scenarios

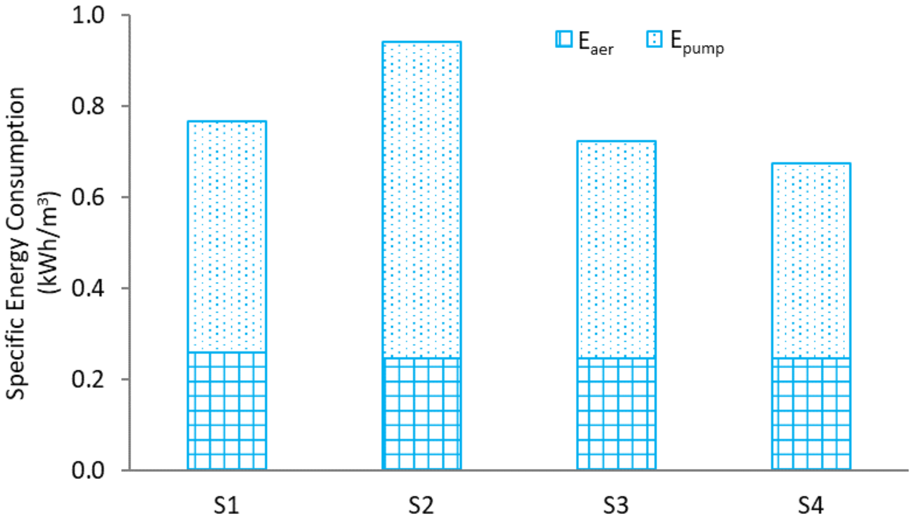

Figure 5. Energy consumption for the aeration and pumping for each scenario

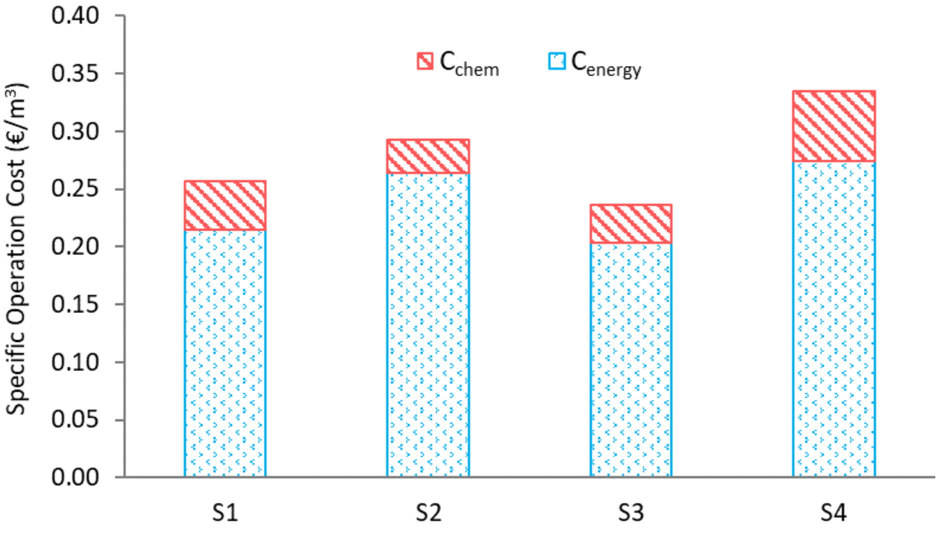

Figure 6. Operation costs of each scenario